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A method of rapid electromagnetic separation of radioactive isotopes

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With 3 figures in the text

ABSTRACT

A method has been developed for rapid, electromagnetic separation of μg quantities of solid substances containing activities. A new ion source equipped with two furnaces has been built. One of the furnaces, containing some mg of carrier, gives the vapour pressure required for the discharge. The other furnace, which contains the active sample, can be connected to the discharge chamber during separation and be heated rapidly up to 1000°C. Separation of Pb^{212} has given 5 % over all efficiency after 7 minutes separation.

Introduction

Most ways of producing radioactive nuclei give a number of active isotopes, even with anisotopic starting material. When these isotopes have equal or nearly equal half lives in isomeric or ground states many problems in nuclear spectroscopy seem to be impossible to solve. Here the electromagnetic separator offers an obvious means of solution in many cases.

At the Nobel Institute a "low intensity, isotope separator" [1, 2] has been in use for some years, mainly in investigations of radioactive gases such as Kr and Xe, but also in connection with other problems in nuclear physics, e.g. production of nuclear reaction targets. At the present time the instrument is used mostly for separation of active solid materials.

The ion currents from the radioactive isotopes in a typical separation are of the order of 10^{-12} A. From this point of view carrierfree amounts would be handled equally well in a mass spectrometer. However, from a practical point of view one is forced, when working with problems such as those discussed here to use ion sources that operate at higher ion currents in order to have larger efficiency and more easily detected reference mass spectrum than in a mass spectrometer. The usual way of handling radioactive substances is, thus, to mix it with a much larger quantity of stable material.

The amount of stable material is settled by the required pressure in the ion source to maintain the discharge. For gases and liquids this can of course be a very small quantity. But for solids one needs a furnace in connection with the ion source in order to have sufficient vapour pressure, and then the amount of charge material needed in the furnace in order to obtain stable running conditions is of the order of mg. Using ion currents of about 10 μA this means a separation time of about 1 hour. If the purpose is to separate stable isotopes, this is no problem as then there is no drawback to charging the furnace with a suitable amount of material. However, when working with approximately carrierfree, radioactive starting material, one is usually forced to dilute the activity with stable material in order to obtain the necessary amount of charge material. An hour is therefore required to separate the activity, which, regarding the amount of material it represents, corresponds to a separation time of only a few seconds.

The following is a description of a procedure which makes it possible to handle very small quantities of material ($\leqslant 1$ μg) including radioactivity. The method involves, in principle, the use of two furnaces in connection with the discharge chamber. The stable material in one furnace is put in mg quantities, and the instrument is put into operation. The other furnace contains the carrierfree, or nearly carrierfree, radioactivity. When the separator is working satisfactory this one is connected to the discharge chamber and heated up. In this way the separation time can be reduced considerably, i.e. to a few minutes.

Thus it should be possible to apply isotope separation to activities with half lives in the range of a few minutes up to an hour. By using only one furnace the more short lived activites cannot be handled with tolerable efficiency.

Some characteristics of the isotope separator

Before a detailed discussion of the new separation procedure and its technical performance it is suitable to make a few remarks about the separator resolving power and efficiency which are intimately connected with gaining a faster separation.

Resolving power

A rough measure of the degree of separation is given by the usual spectroscopic definition regarding those masses resolved having the dispersion between neighbouring isotopesequal to the half-width of the mass distribution. The limit of the resolving power obtained for this apparatus [2] is about 1400. If one assumes the mass distribution for each isotope is triangular, masses below 700 will be fully separated. However, to know the mix in of isotopes one has to measure the distribution from one isotope along the target. This has been done with Kr^{85} [2]. For neighbouring masses the mix in is about 0.1 %, and it decreases to the measurable ratio of about 5×10^{-5} five masses away. To utilize this good separation factor the running conditions and stabilization arrangements of the apparatus have to be very good. It is obvious that this is more easily obtained in a short separation time than in a long one. The figures given above were for a gas, which is usually more easy to handle than solid substances. For these it is even more imperative to have a short separation time in order to be able to use the best resolving power obtainable.

Efficiency

The over all efficiency of an electromagnetic separator is a product of three independent processes. (1) Creation and extraction of the ions; (2) focussing and mass selection; and (3) collection.

For instruments of the type discussed here the second point, that means the ion

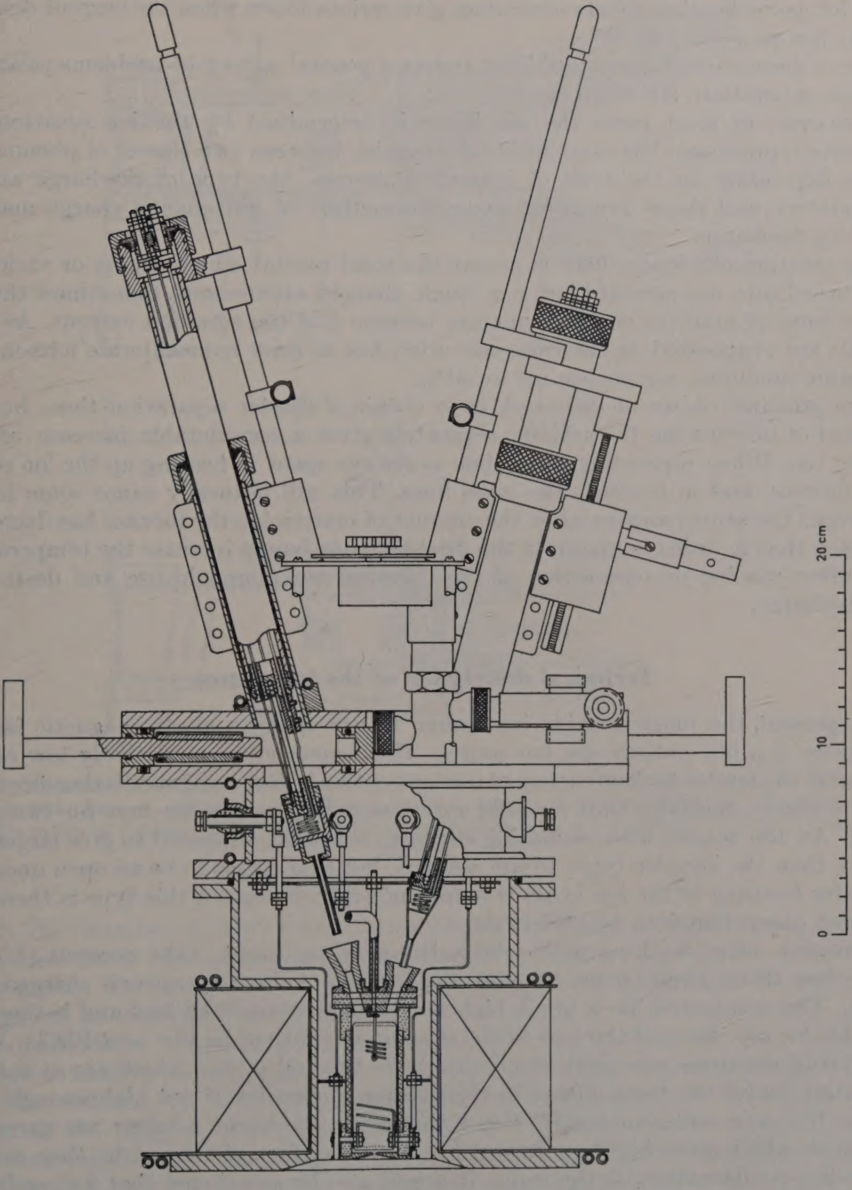

Fig. 1. The ion source.

optics, gives nearly 100 % transmission as no stops are used for resolving power up to the order mentioned above.

The collection consists of direct deposition on a target plate, very often thin Al-foils. For ion currents in the microampere region no sputtering effects give losses;

but, for gases, heating effects sometimes give serious losses when the current densities are as low as a few $\mu A/cm^2$.

For a discussion of these problems and as a general survey of problems related to isotope separation, see reference [3].

However, in most cases the efficiency is determined by the ion creation and extraction processes. Here one has to distinguish between two classes of phenomena: those depending on the type of ionization process, the type of discharge and its parameters, and those depending upon the method of introducing charge material into the discharge.

By creation efficiency, here is meant the total rate at which atoms or molecules are turned into one special kind, e.g. single charged atomic ions. Sometimes the loss in the form of multiple charged ions can become $\frac{1}{3}$ of the total ion current. As most metals are evaporated as chlorides one often has at least monochloride ions in considerable amounts, sometimes up to 50 %.

The principle object of this work is to obtain a shorter separation time, but the method of introducing the activity separately gives a considerable increase of efficiency too. When separating some time is always spent in heating up the ion source and furnace, and in focusing the mass lines. This will naturally cause some loss of material. The same happens when the amount of material in the furnace has decreased so much that in order to maintain the discharge one has to increase the temperature, also often causing decomposition of the chemical compound in use and destroying the discharge.

Technical description of the ion source

At present the most suitable ion source to use with an electromagnetic isotope separator is a low voltage arc ion source. This source gives a relatively low energy spread of the ions at high efficiency. A variant of this is that using oscillating electrons. Such a source, specially built for solid substances, has been in use here for two years [2, 4]. An ion source with oscillating electrons is usually supposed to give larger efficiency than the simpler types of arc sources, but that seems to be an open question. A better focusing of the ion beam is indeed possible. For gases this type is therefore, in most cases, found to be preferable.

However, when working with solid substances one has to take account of other facts than those given above. Suppose the furnace of the ion source is charged with $PbCl_2$. This compound has a much higher vapour pressure than lead and is therefore suitable for use. Most of the ions in the discharge will then be Pb^+ and $PbCl^+$. When oscillating electrons are used, some ions go to the end plates, which are at cathode potential, and if the temperature in the discharge chamber is not high enough most of the Pb^+ ions will condensate. For a simple arc discharge a larger arc current is obtained, which gives higher temperatures on the surface of the anode, thus causing much less condensation on the walls. It might also be mentioned that for oscillating electrons the discharge and its ion densities are much more dependent on the voltage and current parameters than for the simple arc. For these reasons we have always used this latter type of source for solids.

The discharge chamber is built of separate, easily machined parts of stainless steel, molybdenum, pyrophylite and aluminium oxid. These materials give low gasing after having had air in the ion source. At higher temperatures chlorine combines with components in the stainless steel. The chamber and the two furnaces are shown in

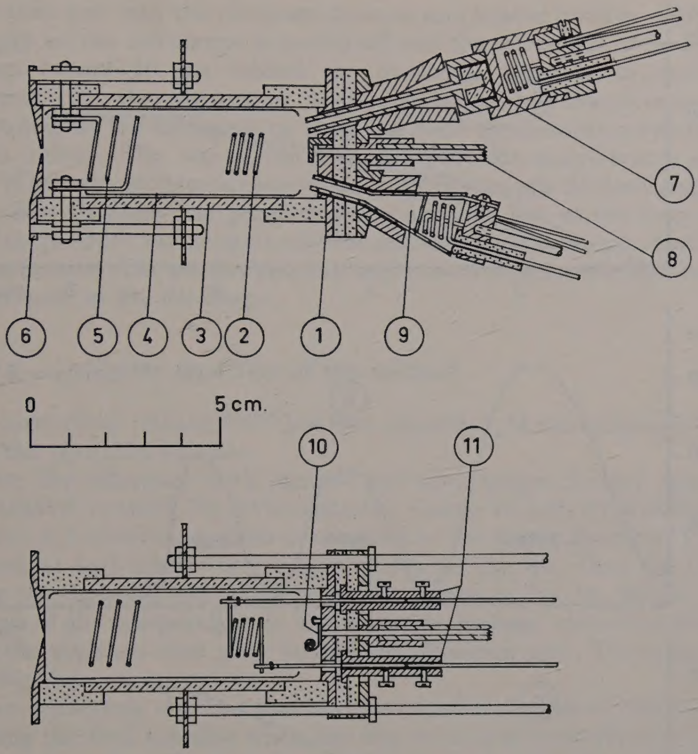

Fig. 2. Schematic drawing of the discharge chamber seen from two perpendicular directions. *1*, Back end plate; *2*, cathode; *3*, insulating wall; *4*, anode; *5*, heating filament; *6*, outlet plate; *7*, stable material furnace; *8*, gas inlet; *9*, platinum furnace; *10*, molybdenum shutter; *11*, cathode holders.

Fig. 2. The chamber, *3*, is a cylinder of insulating materials, with end plates, *1* and *6*, of stainless steel. Inside the insulating cylinder there is a molybdenum tube which is the anode. This arrangement makes it possible to use a discharge of either low voltage arc or oscillating electron type.

The cathode, a tungsten spiral, *2*, is suspended in molybdenum rods which are fastened to holders of stainless steel, *11*. As there has been shown to be a metallic deposit from the cathode, the insulating parts at the cathode holders are in shadow with respect to the tungsten wire. A separate heater is needed in order to obtain higher maximum temperature or to increase the temperature without changes in the power input to the cathodes. Since this heater, *5*, is here placed inside the chamber, the temperature distribution is homogeneous. Both this hot anode and the cathode are fed by direct current because of their accompanying magnetic fields.

The charge, gas and vapour come through the bottom plate, *1*. Over the gas inlet there is a small roof serving as a scatterer. The opening for the stable material furnace is not covered for this furnace is in place during the whole separation time, as is shown in Fig. 2. At the opening for the active material furnace there is a small molybdenum shutter. This will be discussed in detail later. To measure the temperature

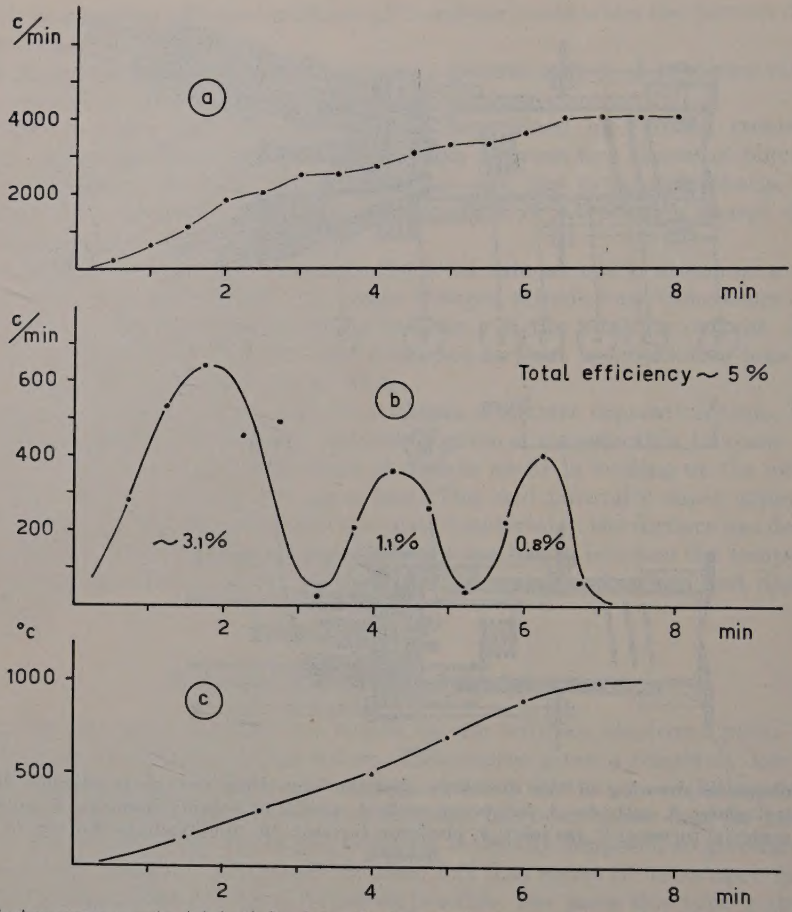

Fig. 3. Test measurements. (*a*) Activity from the target plate versus time. (*b*) Difference in counting rate between successive separation time intervals. (*c*) Temperature of the furnace holder as a function of time.

of the chamber a thermocouple (not shown in the figure) is fastened to the outlet plate, *6*.

The two furnaces are both indirectly heated. The stable material furnace is made of stainless steel with an inner box of graphite or other inert material. The active material furnace is of platinum, chosen for mechanical reasons (this is not a suitable material for all substances). Both furnaces are fastened by long stainless steel rods to brass tubes, see Fig. 1. Conductors go through these tubes for the heating current and thermocouple. Because of the small volume of the tubes and the separate vacuum forepump the furnaces can be put into the main vacuum in one minute without destroying the high vacuum necessary for operation.

Below, the fast separation procedure will be described and some remarks will be made about connected technical problems arising.

Including preheating of the discharge chamber it takes at least half an hour to reach stable operating conditions of the isotope separation. The solution with the active preparate is then put into the platinum furnace and heated until dry. Then the high voltage supply to the ion source is turned off and the furnace is connected into the main vacuum (about 10 cm behind the discharge chamber in order to avoid heating). After this the separation is started again and the platinum furnace is slowly connected to the discharge by means of some mechanical insulating arrangements and a motor. The top of the furnace opens the molybdenum shutter and is in place. When the shutter is opened there is loss through the hole and to the cold platina top. This decrease the pressure for a moment but as the long platina top gets warm the pressure raises again and the activity separation can start by heating the platinum furnace. The shutter plays an important role since the efficiency depends upon the pressure in the discharge.

Test of the method

As a test, carrierfree, 10 hour Pb^{212} has been separated. It was evaporated to dryness as $PbCl_2$ in the platinum furnace.

To measure the efficiency both furnace and then target activity were measured with a scintillation counter. To investigate the change in activity transfer in time of the separation, a β-sensitive counter is mounted in the target chamber. The counting rate measured at half minute intervals is shown in Fig. 3*a*. Thus the difference in the counting rate between successive intervals is given in Fig. 3*b*. To get an approximate measure of the temperature in the platinum furnace, there is a thermocouple mounted in the stainless steel plate behind the platinum part. The temperature rise is given in Fig. 3*c*.

It has been separately checked that no measureable amount of activity leaves the furnace during the first minutes when the top of the platinum furnace is heated by the discharge chamber. Thus, zero time in Fig. 3 is when direct heating of the furnace is started. This is about half a minute after the furnace has been put in connection to the discharge chamber.

The separation referred to above has rather typical figures, although an over all efficiency of 6 % is the best one found. This value refers to activity separated as lead and no account is taken of the chlorides separated. (The efficiency is of course strongly dependent on pressure in the discharge chamber. For a stable running discharge this means slightly more than minimum pressure for plasma discharge.) The remaining activity in the ion source has been measured to check the dependence of efficiency on other processes than the discharge process and neutral vapour leakage through the outlet. In the discharge chamber a very small quantity remains on the walls, but in the furnace there is generally 5–10 % left. This activity may probably be due to lead trapped in the platinum walls.

It has not been possible to completely explain the origin of the peaks in Fig. 3*b*. However, such fine structure has been reproduced in each separation and has been traced back to the evaporation process. During the whole separation the current of stable ions to target was constant and the temperature in the discharge was in equilibrium. One explanation of the occurrence of these peaks might be the following. At a low temperature, $PbCl_2$ evaporates from the furnace, giving rise to the first peak in Fig. 3*b*. At higher temperature $PbCl_2$ dissociates to PbCl, Pb and possibly some lead oxide compounds, thus causing the other peaks. However, $PbCl_2$ is assumed not to dissociate before its boiling point, 950°C [5], and the effect given above could then not occur.

Under the special conditions prevailing here, however, this macroscopic effect, boiling point, may not be applied; there may, for instance, be monolayer effects. This peak phenomenon therefore needs special investigations with systematic changes of, for instance, amounts of carriers and separated substance.

Older measurements [2] with the same apparatus but using the technique of direct mixing of the carrier and the radioactive sample has given 4.5 % over all efficiency for lead. These separations lasted about 1.5 hr. In ref. [2] the efficiency of 8 % given includes the separated amount of $PbCl^+$ ions. In the separation referred to in Fig. 3 we get another 1 % during the time between the 7th and the 13th minute of separation. Thus total 6 % for Pb^+ ions. This means that the method offers the expected increase of efficiency. For the cases of short time available it can be pointed out that about 3 % efficiency is reached in 3 minutes.

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